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Indications of energetic consequences of decoherence at short times for scattering from open quantum systems

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Decoherence of quantum entangled particles is observed in most systems, and is usually caused by system-environment interactions. Disentangling two subsystems A and B of a quantum system AB is tantamount to erasure of quantum phase relations between A and B. It is widely believed that this erasure is an innocuous process, which e.g. does not affect the energies of A and B. Surprisingly, recent theoretical investigations by different groups showed that disentangling two systems, i.e. their decoherence, can cause an increase of their energies. Applying this result to the context of neutron Compton scattering from H_2 molecules, we provide for the first time experimental evidence which supports this prediction. The results reveal that the neutron-proton collision leading to the cleavage of the H-H bond in the sub-femtosecond timescale is accompanied by larger energy transfer (by about 3%) than conventional theory predicts. It is proposed to interpreted the results by considering the neutron-proton collisional system as an entangled open quantum system being subject to decoherence owing to the interactions with the "environment" (i.e., two electrons plus second proton of H_2). [doi:10.1063/1.3595401]

I. INTRODUCTION

Certain features of quantum mechanics seem strange because they contradict the intuitive, and seemingly reasonable, assumptions of classical physics about how the physical world should behave. One of them is the phenomenon of quantum entanglement (QE) — or Einstein-Podolsky-Rosen (EPR) correlations — which has emerged as one of the most emblematic features of quantum mechanics.[1–4] Entanglement lies at the heart of the intriguing complexity of describing quantum many-body systems found in nature.

In most systems, however, one observes an ultrafast decay (or decoherence) of QE, which is usually caused by system-environment interactions. Representing the main barrier to the realization of a quantum computer,[5] decoherence is subject to many experimental investigations and theoretical studies within the frame of dynamics of open quantum systems.[6]

Let A and B be two entangled parts of a system AB in a pure quantum state. Disentangling A and B is equivalent to the erasure of quantum phase relations between A and B and leads to the formation of a mixed quantum state. It is widely believed that this erasure does not affect the properties of the individual subsystems A and B. Most surprisingly, however, recent theoretical investigations by Schulman and Gaveau,[7,8] and by Erez *et al.*[9] and Gordon *et al.*,[10] revealed that disentangling two systems can in fact cause an increase of their energies. In this paper, we propose the connection of

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these theoretical models with the physics of "fast" inelastic scattering processes, and present first experimental results obtained by means of neutron Compton scattering[12] that support them.

In particular, neutron Compton scattering from H_2 molecules, with energy transfers about 19 electron volts, reveals that the individual neutron-proton collision is accompanied by an energy transfer which is significantly larger than predicted by conventional theory. Equivalently, the center-of-gravity of the experimental Compton profile of the proton (H atom) appears to be displaced to positive momenta. As proposed and discussed below, these findings can be understood by considering the neutron-proton collisional system as an open quantum system being subject to decoherence caused by its interactions with the particles of the environment (i.e., electrons and second proton of H_2). In more illustrative terms, one may say that the relevant quantum system becomes "observed" by the environment,[6] thus inducing decoherence.

II. ENERGETIC CONSEQUENCES OF DECOHERENCE

A. The model by Schulman and Gaveau

We begin with a short description of the the general formalism of Schulman and Gaveau.[7,8] A quantum system A with free Hamiltonian H_A makes an elastic collision with a second system B with free Hamiltonian H_B. Let the interaction Hamiltonian (potential) be V_{AB}. The total Hamiltonian is then

$$H = H_A + H_B + V_{AB} \; . \tag{1}$$

Initially, i.e. before the collision, the two particles are not entangled and so we take the complete density matrix $\rho(0)$ to be a product, $\rho(0) = \rho_A(0) \otimes \rho_B(0)$. Subsequent to their collision they become entangled and the exact density operator

$$\rho(t) = U(t)\, \rho(0)\, U^{\dagger}(t)$$

($U(t)$: time evolution operator) is not representable by the product of the individual density matrices $\rho_A(t) = \mathrm{Tr}_B \rho(t)$ and $\rho_B(t) = \mathrm{Tr}_A \rho(t)$; here Tr_i denotes the partial trace over the variables of system i. The existence of this QE follows from first principles within quantum theory. In this context it should be noted that the phenomenon of entanglement is not addressed in conventional neutron scattering theory.[12–14]

Despite this consequence of entanglement, however, it is widely believed that once the particles are separated the quantum correlations associated with the entanglement can be dropped (provided one does not do an experiment of Einstein-Podolsky-Rosen type[1]), simply because measurements of physical quantities of each of the two particles cannot depend on their QE. Thus the replacement $\rho(t) \rightarrow \rho_A(t) \otimes \rho_B(t)$, i.e. the erasure of entanglement, is assumed to be innocuous, as it should not affect the energies of the systems.

The striking result is that this does not hold in general.[7] Putting

$$\Delta\rho(t) = \rho_A(t) \otimes \rho_B(t) - \rho(t)$$

and for a particular form of the interaction Hamiltonian, it was shown that for sufficiently *short* times the following relation holds:

$$\Delta E \equiv Tr[\Delta\rho(t) H] = Tr[\Delta\rho(t)\, V_{AB}] > 0 \; . \tag{2}$$

A detailed derivation of this inequality was presented in a following paper.[8] Moreover, this result was shown to be valid for a large class of potentials, as e.g. for two-body interactions, although it does not hold universally.[8]

In other words, the replacement of the entangled $\rho(t)$ by the non-entangled $\rho_A(t) \otimes \rho_B(t)$, i.e. decoherence of the AB system, necessarily increases the systems's energy, for sufficiently short times.[7,8] This seems highly paradoxical since, as the authors put it: "...losing quantum correlations should not heat the gas. You do not burn your finger because of a partial trace over a density matrix".[7]

One may object that the counter-intuitive result, Eq. (2), violates energy conservation. This, however, is not the case, as the presented discussions[7] showed. It was stressed that, in the situation

contemplated therein, the coupling Hamiltonian must be considered as time dependent, because the physical approach and separation of the particles leads to a time-dependent coupling coefficient. Thus, energy conservation need not apply. It was discussed that the translational degrees of freedom of A and B supply this "additional" energy $\Delta E > 0$.[7] A consistent quantized treatment of the complete collision dynamics that shows this feature has not been given yet.

In the context of our neutron Compton scattering[12] experiment under consideration, the impinging neutron n may be associated with system A, and the struck proton p together with its effective Born-Oppenheimer (BO) potential with system B. The neutron-proton interaction is associated with V_{AB}. Thus Eq. (1) represents an effective Hamiltonian; see also below.

B. The model by Erez *et al.* and Gordon *et al.*

It should be noted that the model of Ref. 7 assumes the appearance of decoherence, as the replacement $\rho(t) \rightarrow \rho_A(t) \otimes \rho_B(t)$ shows, but does not describe the disentanglement process explicitly. Moreover, the environment and the coupling of the two systems to it do not appear explicitly in the model.

Theoretical investigations that address these issues have been presented by Erez *et al.*[9] and Gordon *et al.*,[10] which explore explicitly the aforementioned decoherence process. The focus here is the cooling of two-level model systems (quantum bits) on ultrashort time scales. It is shown that these anomalous non-Markov cooling processes stem from the hitherto unfamiliar coherent quantum dynamics of the system-bath interaction well within the bath memory time. In the course of this analysis the authors also derive a result analogous to Eq. (2), first for brief quantum non-demolition (QND) measurements[9] and later for the general case of any abrupt disturbances, e.g. phase change.[10]

In Ref. 9 the "measurement" (or interaction) of a quantum system S by a "detector" D (i.e a second quantum system) is considered, represented by the interaction $H_{SD}(t)$. The authors study the total Hamiltonian $H_{tot} = H_S + H_B + H_{SB}$ of the system (with Hamiltonian H_S) that interacts with the bath (with Hamiltonian H_B) and is intermittently perturbed by the coupling of the system to the detector:

$$H(t) = H_{tot} + H_{SD}(t) \, . \tag{3}$$

It is essential that the interaction Hamiltonians H_{SB} and H_{SD} do not presuppose the rotating-wave approximation; i.e. energy conservation between the system and the bath or the detector is not imposed, on the ultrashort time scales considered.[9] It was proven in Ref. 10 that an impulsive disturbance of the system always produces an energy increase (termed heating) of the equilibrium state immediately thereafter. This general conclusion was supported by numerical results, for a disturbance of finite, albeit brief, duration.[9] For an ultrashort time interval $0 \leq t \leq \tau$, the authors show the important results

$$\begin{aligned} \langle H_{SB}(0) \rangle \leq 0 \rightarrow \langle H_{SB}(\tau) \rangle = 0 \\ \langle H_{SD}(t) \rangle \; = \; - \langle H_{SB}(t) \rangle \, . \end{aligned} \tag{4}$$

This transfer of energy, associated with a change in the entanglement between the system and the bath, triggers the quantum dynamics that redistributes their mean energy (and entropy). Experimental evidence for this effect in the field spin dynamics studied with nuclear magnetic resonance (NMR) was recently provided.[11]

To make qualitative contact with the experiment under consideration, one may associate the proton p with the quantum system S, the proton's environment (denoted by $\mathcal{E}$ below) with the bath B, and the neutron with D. The aforementioned "additional" energy ΔE (in Ref. 10 denoted by $\delta \langle H_{tot} \rangle^M$ for projective measurements and by $\delta \langle H_{tot} \rangle^\phi$ for an abrupt phase shift) was proven to be positive, $\Delta E > 0$; i.e., a heating of the equilibrium state appears immediately after the external abrupt disturbance.[10] Noting that the NCS process is not a QND process, one may anticipate that ΔE should contain a contribution from the translation energy of the impinging neutron.

III. DETAILS OF COMPTON SCATTERING

In view of the explanations of the preceding section, it should be noted that conventional neutron scattering theory (see e.g. Refs. 13 and 14) is based on the formalism of standard time-dependent first-order perturbation theory (specifically Fermi's Golden Rule), which assumes stationary initial and final scattering states. Thus there is no notion at all of decoherence, which is an inherently time dependent phenomenon.

A. Energy and momentum transfers

We now outline the experimental method. As originally proposed by Hohenberg and Platzman,[15] neutron scattering at high energy and momentum transfers (say, $\hbar\omega > 1$ eV, $q > 20$ Å^{-1}) can be used to directly measure the distribution of momentum $\hbar\mathbf{p}$ of light atoms in condensed matter systems. This experimental method is analogous to the measurement of the electronic momentum distribution through Compton scattering of X- and γ-rays from electrons,[16] or the nucleon momentum distribution via quasi-elastic electron scattering from nuclei,[17] and is known as *neutron Compton scattering* (NCS).[12] Here it is assumed that the scattering is essentially incoherent and the so-called *Impulse Approximation* (IA) applies, and thus single-particle properties are probed.[12,15,16,18] It should be noted that the characteristic time τ_{sc} of a neutron-atom collision, termed "scattering time", is very short. In our experiment, the τ_{sc} of $(p-n)$ collision lies in the sub-femtosecond regime; see below.

From the measured time-of-flight (TOF) spectra, momentum ($\hbar\mathbf{q}$) and energy ($\hbar\omega$) transfers from the neutron to the stuck particle are determined by standard methods; see Appendix. Within the IA, the elastic collision of a neutron with an approximately free atom with mass M and initial momentum $\mathbf{p}$ results in the neutron's lost energy $\hbar\omega$ being transferred to the struck atom:

$$\hbar\omega = \frac{(\hbar\mathbf{q}+\mathbf{p})^2}{2M} - \frac{p^2}{2M} = \frac{(\hbar q)^2}{2M} + \frac{\hbar\mathbf{q}\cdot\mathbf{p}}{M} \\ = \hbar\omega_{rec} + \hbar\mathbf{q}\cdot\mathbf{p}/M \ . \tag{5}$$

The recoil energy $\hbar\omega_{rec} = \hbar^2 q^2/2M$ represents the energy of recoil of a stationary nucleus owing to the collision. Thus scattering from a gaseous sample of such atoms leads to a recoil peak centered at $\hbar\omega_{rec}$, exhibiting a peak width caused by the term $\hbar\mathbf{q}\cdot\mathbf{p}/M$ which represents Doppler broadening. The above energy balance holds exactly in the IA, which however is not completely fulfilled at moderate momentum and energy transfers. In such cases, so-called final state effects (FSE) become apparent. They are caused by environmental forces on the struck particle and can be treated rigorously as a power series in $1/q$.[18] It was shown that FSE cause a shift of the maximum of the recoil peak to *lower* energy than the recoil energy.[12,18] Further theoretical insight is provided by the analytic proof of the existence of this shift in the case of the three-dimensional quantum harmonic oscillator.[19]

In the context under consideration, this point is crucial and thus merits further comments. In the framework of conventional NCS theory, a shift to higher energy-transfer values than $\hbar\omega_{rec}$ would imply that there is a higher energy loss in the scattering process than the IA predicts. Physically this means that the struck atom would be "more free" than an isolated atom floating in space, which of course is meaningless. Hence, deviations from the IA must give peak shifts to less than $\hbar\omega_{rec}$, since they are always caused by the atom not being free, owing to its interaction with other atoms. Thus there is an additional resistance to motion of the struck atom, which manifests as a slightly lower energy transfer than the IA predicts. Summarizing, a peak-maximum shift to higher energies than $\hbar\omega_{rec}$ is impossible within conventional NCS theory.

The measured TOF spectra may be straightforwardly transformed to obtain the associated experimental Compton profiles[12,17,18] of the recoiling atoms, commonly denoted $J(y)$; see Appendix. For isotropic systems, as in our case, this function represents the distribution of the one-dimensional projection $\hbar y$ of atomic momentum $\mathbf{p}$ (before collision) along the $\mathbf{q}$-direction represented by the unit vector $\mathbf{e_q}$. According to the IA,[12,17]

$$J(y) = \int n(\mathbf{p})\,\delta(\mathbf{p}\cdot\mathbf{e_q} - \hbar y)\,d\mathbf{p} \ . \tag{6}$$

Here, $n(\mathbf{p})$ is the momentum distribution of the struck particle with mass M before collision, which is the quantity of interest for theoretical investigations. In the IA, $J(y)$ must be centered at $y = 0$. For further details, see Appendix. The aforementioned conclusion about peak shifts implies that a shift of the peak maximum (or center of gravity) of $J(y)$ to $y_{max} > 0$ values is impossible within conventional theory, since deviations from the IA must displace $J(y)$ to $y_{max} < 0$.

B. Collisional time window

As discussed below (in Sect.V), the ultrashort duration of the $n - p$ collision appears to play an essential role in the interpretation of our experimental observations.

The characteristic time window of the neutron-nucleus collision, often termed scattering time, τ_{sc}, was first obtained by Sears[12,18] as

$$\tau_{sc} \approx \frac{M}{q\sqrt{\langle p_q^2 \rangle}} , \tag{7}$$

where p_q is the projection of particle's momentum $\mathbf{p}$ along $\mathbf{q}$ and the expectation value $\langle ... \rangle$ is taken over the particle's ground state. It is also found that $\tau_{sc} \approx \hbar/\Delta E_q$, where ΔE_q is the width of the dynamic structure factor at q =const. A similar estimate of τ_{sc} can be obtained with the relation

$$\Delta_E \tau_{sc} \geq \pi\hbar/2 \tag{8}$$

derived by Ballentine,[20] where τ_{sc} may be interpreted as the shortest time required for the state vector $|\psi(t)\rangle$ to become orthogonal to the initial state $|\psi(0)\rangle$, and Δ_E represents a standard measure of the statistical spread of energy in the state (e.g. the standard deviation).

In our NCS experiments, the characteristic time of the neutron-proton scattering process is of the order: $\tau_{sc} \sim 10^{-16} - 10^{-15}$ s.

The notion "scattering time" may be, in some circumstances, misleading. A better way to illustrate the physical meaning of τ_{sc} is the following: First, note that the time interval during which the neutron and the nucleus are in physical proximity may be even shorter, as the following estimate shows. For example, a neutron with kinetic energy $E_0 \approx 10$ eV will pass a distance of 10^{-5} Å, which is about the range of the strong nuclear force, in a much shorter time, i.e. $10^{-19} - 10^{-20}$ s. However, this is not in conflict with the above estimate, for the following important reason: τ_{sc} gives a measure of the length of the time interval during which the neutron-proton collision (owing to the strong nuclear force with range ca. 10^{-5} Å) may occur.

Thus, in the light of the "attosecond double-slit" experiment by Paulus and co-workers,[21] the scattering process may be viewed as a quantum interference process over the time interval τ_{sc}. This conceptual viewpoint (i.e., interference-in-time) is also in line with the results obtained from a temporally diffracted beam of slow Cesium atoms, using a Young-slit-type interferometer in the time domain.[22]

IV. RESULTS

Figs. 1 and 2 show the Compton profiles $J(y)$ of D and H obtained from measurements of gaseous H_2 and D_2 on the eVS/Vesuvio NCS spectrometer at the neutron spallation source ISIS. These experimental data were obtained by transforming to y-space the TOF spectra measured as described above and in the Appendix.

We have analyzed the sensitivity of the peak positions of $J(y)$ in y-space to calibration uncertainties and found that the results in y-space are reliable to better than 0.5 Å^{-1}. This estimate is supported by the small scattering of the positions of the individual-detector $J(y)$ curves shown in all four Figs. 1(a), 1(b), 2(a), and 2(b). The most important findings are:

(A) The peak maxima (or the centers-of-gravity) of the D Compton profiles are found to be at $y_{max} \leq 0$ as conventionally expected, for all momentum transfers investigated (Fig. 1(a) and 1(b)). The IA appears to be well obeyed for high momentum transfers. In these experiments, the D-D bond is not broken (see below). The Compton profiles (a) and (b) are in full agreement with the

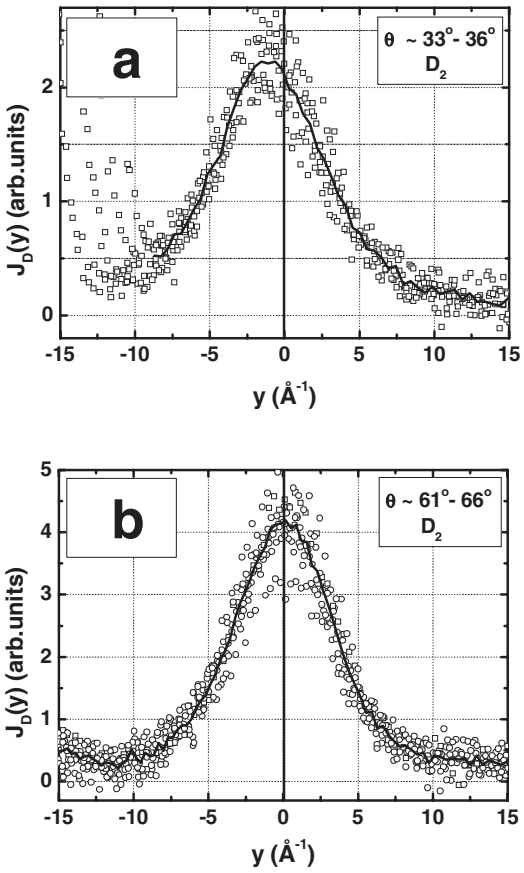

FIG. 1. Experimental Compton profiles $J_D(y)$ of D in D_2 gas at 44 K and 10 bar. **(a)** $J_D(y)$ measured with one block of 8 detectors, at the lowest ($\theta_{low} \sim 33^\circ - 36^\circ$) scattering angles available. The results clearly exhibit conventional FSE for low momentum transfers. **(b)** $J_D(y)$ measured with one block of 8 detectors, at the highest ($\theta_{high} \sim 61^\circ - 66^\circ$) scattering angles available presently. In all experiments, the D-D bond is not broken after scattering; see the text.

well-established results of Andreani *et al.*[23] Therefore they also represent a confirmation of both the correct working of the present configuration of the eVS/Vesuvio spectrometer and our data analysis, and a baseline for comparison with our results for H, measured and analyzed in the same way.

(B) The H Compton profiles obtained at low scattering angles are centered at $y_{max} \leq 0$ as conventionally expected and experimentally observed earlier;[23,24] see Fig. 2(a). In contrast, the H Compton profiles measured at θ_{high} are centered at momentum values $y_{max} > 0$, thus contradicting conventional theory; see Fig. 2(b). Equivalently, the actual energy transferred in the collision is larger (by about 3%) than the IA predicts.

The magnitude of this effect may be estimated as follows: From Eq. (6), the excess energy transfer is

$$\delta E = \hbar^2 q\, y_{max}/M \tag{9}$$

and thus the ratio R of δE to the recoil energy is $R \equiv \delta E/\hbar\omega_{rec} = 2y_{max}/q$. For H, $m \approx M$ and $q \approx k_1 \tan\theta$, where k_1 is the wavevector of the scattered neutron. From the observed shift of $J_H(y)$ at high q we estimate $y_{max} \approx 1.5\,\text{Å}^{-1}$, which amounts to an excess energy $\delta E \approx 590$ meV. Therefore one obtains $R \approx (2 \cdot 1.5)/(48.6 \cdot \tan 63^\circ) = 0.03$. This is interpreted to represent the quantity ΔE of Eq. (2), i.e. $\Delta E = \delta E$.

(C) One may wonder whether less detailed scattering data (e.g. energy-integrated Compton profiles, the determination of which would require less effort) could be capable of showing the new result (B) as well. This seems not to be the case, as the following example indicates. The

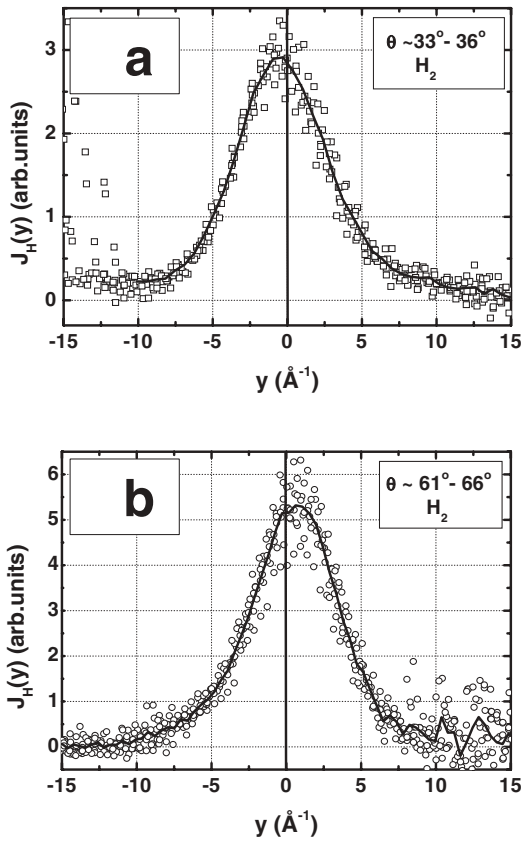

FIG. 2. Experimental Compton profiles $J_H(y)$ of H in H_2 gas at 41 K and 10 bar. The instrumental setup was the same as for the experiment with D_2; see the text and Fig. 1. The results **(a)** at θ_{low} are in agreement with the corresponding results of Andreani *et al.*,[23] in which conventional FSE are visible in the experimental Compton profiles $J_H(y)$. The H-H bonds are not broken after scattering. In contrast, the results **(b)** at θ_{high} (with $q \sim 95$ Å^{-1} and $\hbar\omega \sim 19$ eV) are centered at momentum values $y_{max} > 0$. Here the H-H bonds are broken after scattering. We note that results from detectors at $\theta > 50°$ were not reported in Refs. 23 and 24.

energy dependence of the above angle-resolved scattering results should be compared with those of a corresponding experiment measuring "total scattering", i.e. integrated over all angles. As visual inspection of Figs. 1(a) and 1(b) indicates, the integrated Compton profile of D, $\overline{J_D}(y)$, should remain centered at $y_{max} < 0$ and thus it could be interpreted to exhibit conventional FSE. From Figs. 2(a) and 2(b) one may conclude that the integrated Compton profile of H, $\overline{J_H}(y)$, should be centered at $y_{max} \approx 0$, and thus it could be interpreted to exhibit no FSE at all within present experimental error, which of course would not represent a contradiction to conventional theory. In this context, it may be noted that the determination of the total scattering cross-sections of H and D (and the experimental test of the associated sum rules[13,14] of conventional theory) are beyond the scope of the present paper.

V. DISCUSSION

Observation (B) stands in strict contrast to conventional NCS theory for the reasons described above. It may, however, find a plausible explanation within the framework of open quantum systems, as we propose in the following.

Before collision, the proton is assumed to be bound in an effective BO potential (see Eq. (1)) which, in the usual theoretical picture,[12] is related to the "environment" $\mathcal{E}$ (the two electrons and the second proton of H_2); the three quantum systems n, p and $\mathcal{E}$ are not entangled. The $n - p$ collision creates QE between the two particles, indicated by $\rho_{np}(t)$, see Sect. II. Furthermore, within the time

window τ_{sc}, the struck proton collides and/or strongly interacts with its environment $\mathcal{E}$, causing a complex many-body quantum dynamical process. These interactions create new entanglement between the involved charged quantum systems p and $\mathcal{E}$, which then naturally leads to decoherence of the neutron-proton system under investigation. Thus one may write

$$\begin{aligned} \rho_n(0) \otimes \rho_p(0) \otimes \rho_{\mathcal{E}}(0) &\rightarrow \rho_{np}(t) \otimes \rho_{\mathcal{E}}(t) \\ &\rightarrow \rho_n(t') \otimes \rho_{p\mathcal{E}}(t') \rightarrow \ldots \end{aligned} \tag{10}$$

for $0 \leq t < t' \leq \tau_{sc}$. The third step of this dynamical chain of events is due to the "measurement" of p by the environment $\mathcal{E}$. (Later on, at time t'', and after equilibration of the highly excited system "p plus $\mathcal{E}$", a non-entangled state $\rho_p(t'') \otimes \rho_{\mathcal{E}}(t'')$ may occur again.)

For our purposes, it is important to clarify the physical cause of decoherence in the actual experimental context. To do so, we note that the $p - n$ scattering time related to our NCS experiment turns out to be of the *same order* as the characteristic time of electronic motion and/or charge redistribution, $\tau_{el.motion}$, following the violent excitation and/or electronic shake-up of $\mathcal{E}$, both lying in the attosecond time range:

$$\tau_{sc} \sim \tau_{el.motion} \tag{11}$$

(see also below). These excitations are caused by the sudden movement of the struck proton which, in our experiments, has a mean kinetic energy and an energy spread of several electron volts after the collision, which are of the order of the separation of the Born-Oppenheimer molecular electronic levels (see below for numerical details). Under such conditions, neither the adiabatic nor the sudden approximation[25] can be applied, and thus the "environment" $\mathcal{E}$ appears to play an active role in the quantum dynamics of the $p - n$ system. Additionally, neither can $\mathcal{E}$ be considered here as a memory-less (Markov) bath, nor is it completely "frozen" and does not participate in the dynamics. In more illustrative terms, $\mathcal{E}$ is "observing" or "measuring" the $p - n$ system. This leads to disentanglement and decoherence, which may be expected to have a similar characteristic time.[26]

Obviously, the relation (11) holds only for a restricted range of neutron energies (e.g. those of our experiment), and thus is certainly not valid in conventional thermal and/or cold neutron scattering.

That $\tau_{el.motion}$ lies in the sub-femtosecond time scale has been confirmed by attosecond laser spectroscopy. E.g., Goulielmakis *et al.*[27] reported the real-time observation of valence electron motion in Kr atoms. Moreover, after ionization, the electron-ion pair was demonstrated to constitute an entangled system and its attosecond decoherence was probed.

As mentioned above, this physical picture is not in line with conventional NCS theory, which assumes that single-particle properties are measured, the environment plays no active part in the dynamics, and only stationary scattering states are taken into consideration.

That the effect discussed here is observed for H_2 but not for D_2 can be understood by noting that, under our experimental conditions, molecular dissociation occurs for H_2 but not for D_2. At the recoil energy, H_2 dissociation occurs at approximately $q > 68\,\text{Å}^{-1}$, a condition readily met at θ_{high} where $q_{rec} \sim 95\,\text{Å}^{-1}$ (and corresponding $\hbar\omega_{rec} \sim 19$ eV). In contrast, dissociation in D_2 would occur at about $q > 96\,\text{Å}^{-1}$, which is much higher than the available $q_{rec} \sim 60 - 65\,\text{Å}^{-1}$ at θ_{high}.[24] Consequently, at θ_{high} the neutron-H scattering probes final unbound states in the continuum, whereas neutron-D scattering takes place in the physical space of bound states only. The irreversible electronic shake-up caused by H-H bond breaking should exhibit quite different dynamics from the excitation of bound states of H_2 or D_2. Thus it is not surprising that the decoherence mechanisms in these two cases turn out to cause qualitatively different effects during the attosecond time window τ_{sc}.

Needless to say, the complex five-body dynamical problem (i.e. neutron plus two nuclei plus two electrons) has no analytic solution, and a proper quantitative theoretical model of it is not available thus far. However, first theoretical-numerical investigations of the proton momentum distribution $n_H(\mathbf{p})$ in condensed systems, in relation with NCS processes, have been recently initiated.[28]

VI. CONCLUSION AND ADDITIONAL REMARKS

Generally speaking, the results presented above suggest that the relevant theoretical framework of "fast" collisional processes is that of non-unitary dynamics of open quantum systems.[6] Moreover, decoherence seems to play a more prominent role here which has not been realized so far. The quantitative theoretical analysis of the new results remains a challenge to modern theory.

The complex dynamics summarized in relations (10) merit further comments. Decoherence is the focus of very active research on the foundations of quantum theory.[6] Since 1970, various pioneering theoretical investigations of related processes showed that complete information about a particle (here: struck proton) is carried away into the "environment" by interacting microscopic objects and/or a thermal bath. The corresponding information transfer should be considered irreversible, at least in the sense that the "lost" information cannot practically be retrieved. The reader interested in the historical development of the fundamental theory of decoherence may refer to Refs. 29.

One may wonder why the dynamics of open quantum systems should be relevant for the system "neutron plus proton of H_2", although "neutron plus H_2 molecule" (in gas phase) is a closed system (in the theoretical frame of non-relativistic quantum mechanics.) Here it is instructive to refer to the aforementioned investigation of krypton atoms by Goulielmakis *et al.*[27] applying attosecond laser-spectroscopic techniques. As a result, even this monoatomic system was found to exhibit short-lived entanglement and decoherence, which are typical phenomena of open quantum systems (see above).

Owing to the fact that the timescale of the considered scattering process and of electronic motion are of the same order, i.e. both being in the attosecond regime, we anticipate the above phenomenon to be a starting point for more sophisticated time-dependent studies in atoms (like e.g. ion-electron decoherence accompanying electron extraction from an atom with an attosecond laser pulse) and in molecules (e.g. chemical bond cleavage caused by collisions of other probe particles with molecules).

The general character of the decoherence effect of Refs. 7–9, and 10 leads one to speculate that the new findings presented in this paper might be of relevance for quite different scattering processes (e.g., electron-molecule scattering, proton-nucleus scattering of high energy physics, etc.).

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APPENDIX

We present here a concise description of the measurements of TOF spectra and the associated Compton profiles.

Our NCS experiments were done with the electron-volt spectrometer eVS/Vesuvio at ISIS (Rutherford Appleton Laboratory, UK), which applies the time-of-flight (TOF) technique to determine the energy and momentum transfers to the neutron during each scattering event. The TOF for each detected neutron is given by

$$t = \frac{L_0}{v_0} + \frac{L_1}{v_1} + t_0 \,. \tag{A.1}$$

Here L_0 is the distance from the source to the sample, L_1 that from sample to detector. v_0 and v_1 are the velocities of the incident and scattered neutron, respectively. The detector is at scattering angle θ. t_0 is a small time offset due largely to electronic delays. We used values for L_1 and θ for each individual detector measured with steel rules and a purpose-built protractor.[30]

This spectrometer is a so-called "inverse geometry" instrument meaning that the final velocity v_1 of the neutrons contributing to the TOF spectra is fixed, whereas v_0 varies; for details cf.[30,31] The final neutron energy is fixed at $E_1 = 4906$ meV (the resonance energy of Au-197 used as analyzer foil[32]), corresponding to velocity $v_1 = 3.064 \times 10^4$ m/s and wavevector $k_1 = 48.6\,\text{Å}^{-1}$.

Noting that the geometric quantities L_0, L_1 and θ can be determined by appropriate methods, it follows from Eq. (A.1) that to each t corresponds to an initial velocity v_0 and thus to an energy transfer

$$\hbar\omega = \frac{(\hbar k_0)^2}{2m} - \frac{(\hbar k_1)^2}{2m} \tag{A.2}$$

For the corresponding momentum transfer $\hbar q$ from the neutron to the struck atom, $\hbar\mathbf{q} = \hbar\mathbf{k_0} - \hbar\mathbf{k_1}$,

$$q = \sqrt{k_0^2 + k_1^2 - 2k_0k_1\cos\theta}\ . \tag{A.3}$$

Hence, for each value of t in an experimental TOF spectrum recorded at a particular detector, the associated momentum ($\hbar\mathbf{q}$) and energy ($\hbar\omega$) transfers from the neutron to the struck particle are uniquely determined. For scattering from H, q and ω vary considerably over the range of the recoil peak, especially for high scattering angles.

For scattering from *free* atoms with *zero* initial momentum, $p = 0$, conservation of kinetic energy and momentum in an elastic neutron-atom collision yield the kinematic relation

$$\frac{v_1}{v_0} = \frac{k_1}{k_0} = \frac{\cos\theta + \sqrt{(M/m)^2 - \sin^2\theta}}{M/m + 1} \tag{A.4}$$

which holds in both quantum and classical mechanics. Here m and M are the masses of the neutron and struck nucleus, respectively. This physically corresponds to neutrons detected at the center of the measured recoil peak. For neutron scattering from H, putting approximately $m = M$, it follows that $k_1/k_0 = \cos\theta$.

From the measured time-of-flight (TOF) spectra one determines the dynamic structure factor,[13] which in the case of the IA takes the simple form[12]

$$S(\mathbf{q},\omega) = \int n(\mathbf{p})\,\delta(\hbar\omega - \hbar\omega_{rec} - \hbar\mathbf{q}\cdot\mathbf{p}/M)\,d\mathbf{p}\ . \tag{A.5}$$

$n(\mathbf{p})$ is the momentum distribution of the struck particle before collision. The delta function incorporates Eq. (6).

To compare results obtained with different detectors (and/or to improve counting statistics), one usually applies the so-called West- or y-scaling,[17,18] which, for isotropic samples, amounts to the following. In the IA can be shown that, for each kind of struck particle of mass M, the two variables ω and q are uniquely coupled through a new scaling variable with the physical dimensions of momentum, commonly denoted by $\hbar y$ (or simply y) and defined by

$$\hbar y = \frac{M}{\hbar q}\,(\hbar\omega - \hbar\omega_{rec})\ , \tag{A.6}$$

which simplifies the dynamic structure factor to[12]

$$S(q,\omega) = \frac{M}{\hbar q}J(y), \quad \text{with} \quad J(y) = \int n(\mathbf{p})\,\delta(\mathbf{p}\cdot\mathbf{e_q} - \hbar y)\,d\mathbf{p}\ . \tag{A.7}$$

$J(y)$ is the Compton profile, which gives the distribution of the one-dimensional projection $\hbar y$ of atomic momentum $\mathbf{p}$ (before collision) along the $\mathbf{q}$ direction represented by $\mathbf{e_q}$, i.e. $|\mathbf{e_q}| = \mathbf{1}$. This scaling property of the IA implies that all detectors (at various scattering angles) should yield the same Compton profile $J(y)$.[17,18] This quantity, and the associated $n(\mathbf{p})$, are widely believed to depend on the effective Born-Oppenheimer potential $V(r)$ of the struck particle.[12] For a harmonic $V(r)$ the associated $J(y)$ is Gaussian.

Our measurements on H_2 and D_2 gas were made using an all-aluminium pressure cell with parallel internal surfaces perpendicular to the incoming beam. The mounting of the cell within the cryostat ensured reproducibility of the sample position between experiments to better than about 2 mm. The spectrometer configuration was identical in the two experiments. Exactly the same analysis was applied to each data set.

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